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Advanced Laser-Comptin Gamma-ray Sources for Nuclear Materials Detection, Assay and Imaging

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Advanced Laser-Compton Gamma-ray Sources for Nuclear Materials Detection, Assay and Imaging

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Highly-collimated, polarized, mono-energetic beams of tunable gamma-rays may be created via the optimized Compton scattering of pulsed lasers off of ultra-bright, relativistic electron beams. Above 2 MeV, the peak brilliance of such sources can exceed that of the world's largest synchrotrons by more than 15 orders of magnitude and can enable for the first time the efficient pursuit of nuclear science and applications with photon beams, i.e. Nuclear Photonics. Potential applications are numerous and include isotope-specific nuclear materials management, element-specific medical radiography and radiology, non-destructive, isotope-specific, material assay and imaging, precision spectroscopy of nuclear resonances and photon-induced fission. This review covers activities at the Lawrence Livermore National Laboratory related to the design and optimization of mono-energetic, laser-Compton gamma-ray systems and introduces isotope-specific nuclear materials detection and assay applications enabled by them.

Keywords: gamma-rays, nuclear resonance fluorescence, laser-Compton scattering.

1. Introduction

For more than a decade, the Lawrence Livermore National Laboratory (LLNL) has been developing and advancing the state of the art with respect to laser-based Compton x-ray and gamma-ray light sources. In recent years, work has concentrated on the optimization of laser-Compton gamma-ray sources with respect to size, flux and bandwidth with a specific aim of creating field-worthy, mono-energetic gamma-ray systems for materials detection, assay and imaging applications. Mono-energetic gamma-rays can efficiently excite the electromagnetic resonances of the nucleus and produce nuclear resonance fluorescence (NRF). NRF transitions depend upon the number of protons and neutrons in the nucleus and are thus unique fingerprints of each isotope. By monitoring the attenuation of gamma-ray photons due to the excitation of NRF transitions, one may determine the presence, amount and distribution of specific isotopes within arbitrary objects. Additionally, NRF transitions for actinides of interest to the nuclear power and nuclear security communities occur at photon energies that are highly penetrating, e.g. in the 1 MeV and 4 MeV spectral range. It is therefore possible to not only assay and detect materials with laser-Compton gamma-ray sources but to do so in the presence of appreciable shielding or inside of thick objects. Possible applications enabled by next generation laser-Compton gamma-ray sources include rapid (milliseconds) detection of concealed nuclear material, high precision (better than 100 parts per million) non-destructive assay of spent nuclear fuel assemblies, isotope-specific, high-resolution (less than 10 micron spatial resolution) 3D imaging of nuclear materials in existing waste containers and waste processing streams.

2. Laser-Compton Scattering

Compton scattering of laser photons from relativistic electrons was first demonstrated in 1965.[1] In that experiment a giant pulse ruby laser interacted with 6 GeV electrons and created approximately 8 upshifted photons per laser pulse. In the years following this demonstration, laser Compton scattering was used as a diagnostic of electron beam quality in advanced accelerators. In its simplest configuration, laser light is incident head on with the electron beam and the on axis, upshifted photons have an energy equal to $4\gamma 2E_l$, where γ is the normalized energy of the electron and E_l is the incident photon energy. By monitoring the spectrum of the upshifted photons, one may learn about the energy spread the electron beam. In the 1990's a renaissance in laser-Compton scattering arose from the ultrafast materials community which used the process to produce short duration bursts of x-rays, typically of a few 100 fs to few ps in duration.[2,3] In order to reduce the duration of the resulting x-ray pulse in these systems, the laser was often incident at right angles to the electron beam direction. While these sources produced short duration x-rays, they also produced relatively broadband x-rays (>10% ΔE/E), were relatively inefficient and the up-scattered photon energy was only half of that from a head on collision. Fundamentally the efficiency of laser Compton scattering is limited by the small magnitude of the Thomson cross section (~0.6 barns) and the inability of electron beams to be focused to spots on par with minimum laser spot dimensions. In 2004, LLNL scientists recognized [4] that the Compton scattering brilliance should increase rapidly as a function of electron beam energy and beam quality. To first order this occurs because at higher electron beam energy it is possible to overcome electrostatic repulsion and focus the electron bunch to smaller spot dimensions. Roughly, the electron spot dimension is proportional to its beam energy and thus the peak brilliance (photons/sec/0.1%BW/mrad2/mm2) of the laser-Compton source increases as function of electron beam energy somewhere between 2nd and 4th power. This rapid increase in peak brilliance is illustrated in Figure 1 and is in stark contrast to the trends of alternative sources, such as large-scale synchrotrons. In the nuclear excitation region above 100 keV, the peak brilliance of 3rd generation

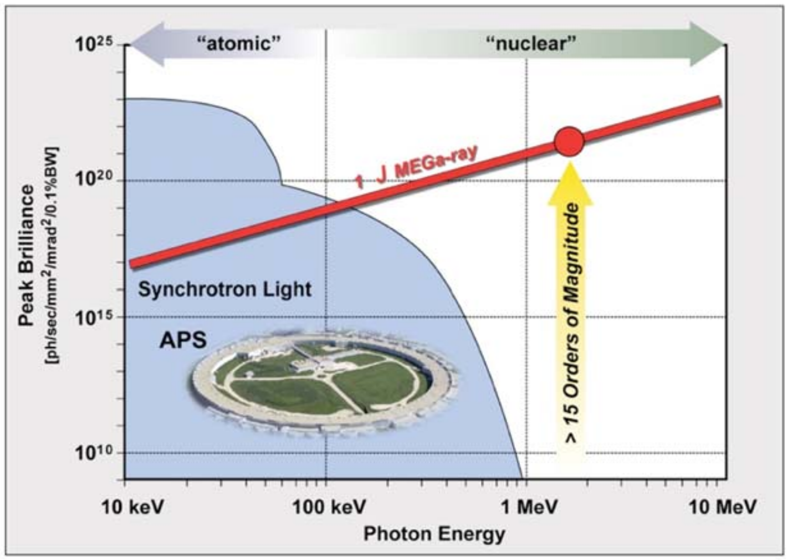

Figure 1. Peak brilliance of a laser-Compton light source relative to the APS synchrotron.

synchrotrons decreases faster than exponentially. Above 2 MeV, the peak brilliance of an optimally-configured, laser-Compton source can exceed that of the largest synchrotrons by more than 15 orders of magnitude. It is important to note that for many nuclear applications and especially for those related to nuclear materials management and detection, it is the bandwidth of the Compton source and not the pulse duration or brilliance that is of foremost importance.

The optimization of laser-Compton scattering to produce narrowband gamma-rays involves a different approach to machine design than that pursued for short duration x-ray sources. The bandwidth of the Compton source is driven by three effects; the energy spread of the electron bunch, the bandwidth of the laser photons and the spread due to the angle correlation in the interaction region. Bandwidth can be minimized with high quality (low emittance) electron beams, few-ps or longer laser pulses and optimized laser-electron interaction geometries. Fractional bandwidths of ~10^{-3} ΔE/E or ~2 orders of magnitude less than that demonstrated from short-duration, laser-Compton x-ray sources are possible with careful design.

3. Excitation of Nuclear Resonance Fluorescence

Laser-Compton gamma-ray sources can enable "Nuclear Photonics", that is the photon-based manipulation and study of the nucleus. In particular narrow-band sources can efficiently excite nuclear resonance fluorescence (NRF). While the width of NRF transitions at room temperature is very narrow (typically 10^{-5} to 10^{-6} ΔE/E), selective excitation is possible with an optimized, 10^{-3} ΔE/E bandwidth laser-Compton source. Furthermore, NRF cross sections of interest are large compared to background and often occur within the max-transparency window for most materials (see Figure 2).

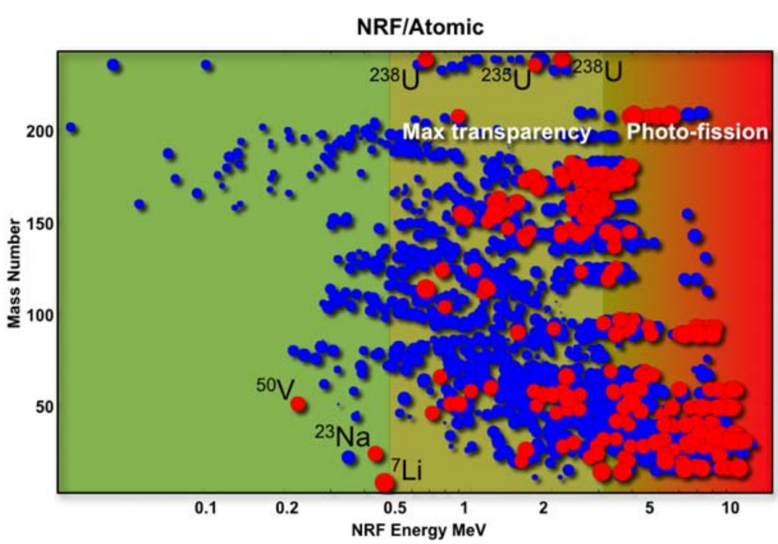

Figure 2. Scatter plot of relative NRF cross sections as a function of mass number and photon energy. Dot size is proportional to cross section magnitude. Red dots are larger than the atomic

background at the particular energy.

Laser-Compton excitation of NRF can be used in “reflection” or in “transmission” to determine the presence or absence of a particular isotope. In reflection, one illuminates the object in question and looks for the characteristic NRF relaxation radiation which is emitted into 4π. In transmission one looks for the absence of resonant photons in the transmitted beam. Besides being intrinsically less susceptible to clandestine attempts to obscure signals, transmission based systems can also provide quantitative assay and high resolution spatial information regarding the isotopic content of the object. In transmission the primary issue is low angle Compton scattering which can create new photons at the resonance energy and degrade measurement accuracy. It has been numerically shown that this problem is alleviated for sufficiently narrowband and collimated laser-Compton sources. [5]

As an example let us consider a standard nuclear fuel rod containing isotopic defects which include variations in density and enrichment (see Figure 3). [6]

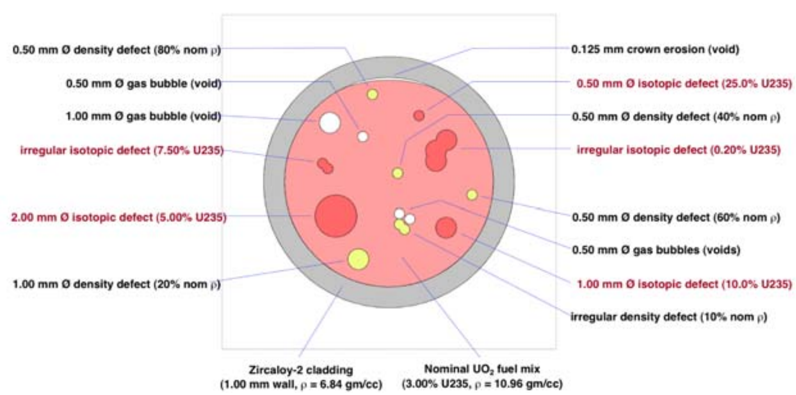

Figure 3. Uranium oxide fuel rod model.

As illustrated in Figure 4, the Bremsstrahlung image can identify the density defects but misses the enrichment variations. However an image obtained with ^{235}U-resonant, 1733-keV gamma-rays is able to identify the location and magnitude of the enrichment variations. Separate analysis [6] suggests that high-flux, narrowband, laser-Compton systems currently being constructed will be able to assay nuclear fuel assemblies with better than 100 ppm accuracy per isotope of interest.

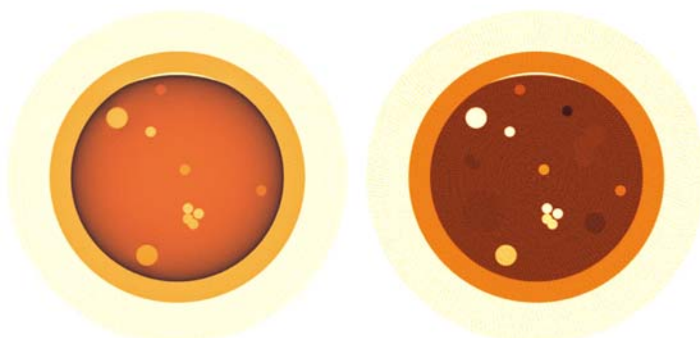

Figure 4. Simulated Uranium oxide fuel rod images with 2 MeV Bremsstrahlung (left) and 1733 keV laser-Compton gamma-rays (right). The simulations were performed with a modified version of LLNL's Monte Carlo code – COG that includes NRF transitions and their line shapes.

4. Detector Systems for Laser-Compton Sources

Transmission based detection and assay of materials with narrow-band, laser-Compton gamma-rays is fundamentally an issue of observing the narrow-band (few eV wide) removal of resonant photons from the much wider (~1000 eV wide) spectrum of the interrogating laser-Compton beam. The detection and measurement of this "notch" in the transmitted spectrum is well beyond the ~1000 eV resolution of today's best single photon counting, gamma-ray spectroscopic techniques. Furthermore the high flux per pulse characteristics of laser-Compton sources is ill-matched to the single-photon-counting operation of traditional high-resolution gamma-ray detectors, e.g. high purity germanium (HPGe). To overcome these issues, a new, calorimetric, dual-isotope notch observation (DINO) detector arrangement was developed and patented (US8369480 B2) by LLNL. Schematically DINO detector arrangement is shown in Figure 5.

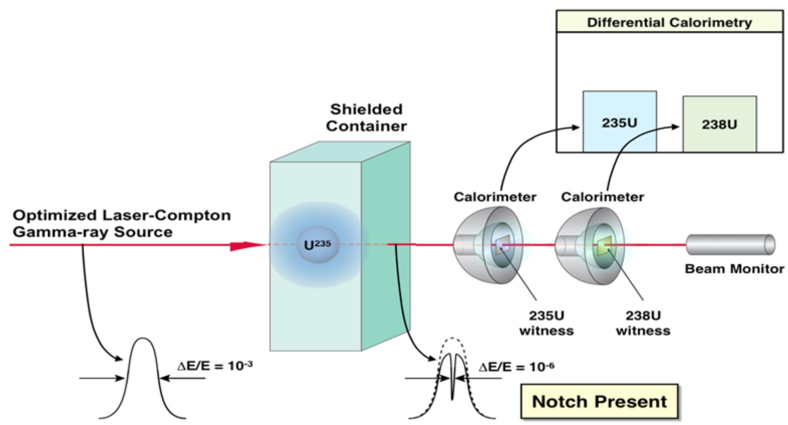

Figure 5. Schematic of the Dual Isotope Notch Observation (DINO) detector arrangement.

In the DINO arrangement illustrated in Fig. 5, the beam is transmitted through the interrogated object and interacts with two "witness" samples before being collected by a beam monitor. The witness samples are composed of two isotopes of the element that is sought in the interrogated object, e.g. ^{238}U and ^{238}U if interrogating a spent fuel rod. The first witness is composed of the isotope for which the laser-Compton beam may contain resonant photons. When the transmitted beam interacts with this witness Delbruck, Rayleigh, Thomson, Compton and NRF scattering will take place. On the other hand illumination of the second witness will only produce Delbruck, Rayleigh, Thomson and Compton scattering. Because the magnitudes of the non-resonant scattering cross sections (Delbruck, Rayleigh, Thomson and Compton) do not change rapidly with nucleon number, the scattering from the second witness provides an accurate measure of the non-resonant scattering present in the first witness sample. Thus the properly normalized difference of the integrated scattered energy from both samples is proportional to only the amount of NRF scattered by the resonant witness. The more resonant material that is present in the object, the more resonant photons that will be removed from the transmitted laser-Compton beam and the less difference in the normalized scattered energy from the two witness materials there will be. By placing known quantities of the desired isotopic material in the beam, it is possible to create an absolute calibration of the calorimetric difference between the scattered radiation from the two witnesses and thus to determine not only the presence of material in the interrogated object but also the precise amount of material in the object. Unlike reflection based detection systems, the DINO arrangement can tell by the lack of sufficient photons on the beam monitor if the object has been two heavily shielded to determine the presence of the desired isotopic material. This is an important attribute for nuclear materials security applications. Furthermore the DINO detector arrangement does not require the use of single photon counting detectors and thus can take full advantage of the high-photon-number per pulse characteristics of optimized laser-Compton sources.

The design metric of importance for NRF-based interrogation with laser-Compton sources is the ratio of the number of photons from the source that are resonant with the desired material per unit time (i.e. the signal) to the number of photons that are not resonant (i.e. the noise). This is proportional to the specific spectral density (SSD) of the source or the number of photons per second per unit bandwidth divided by the fractional bandwidth of the source. As illustrated in Figure 6, new detection, assay, imaging and science capabilities are enabled as the SSD of the source increases.

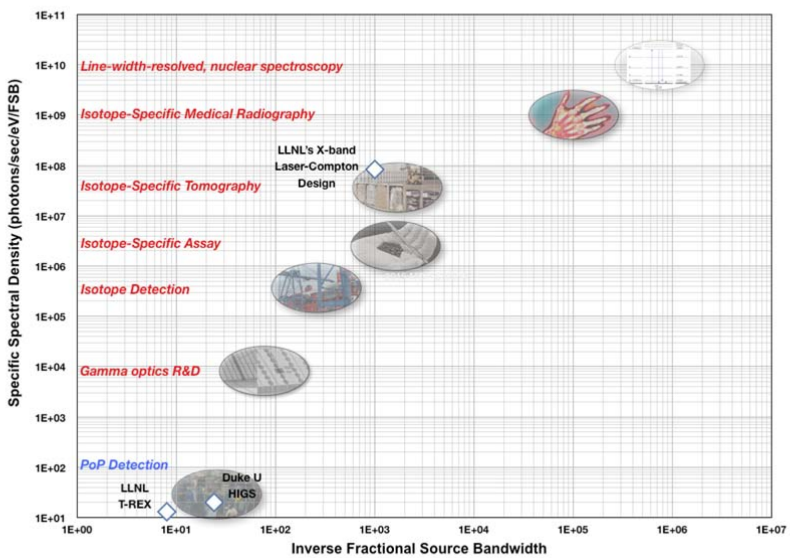

Figure 6. Potential applications enabled by increased source specific spectral density (SSD).

5. Optimized Laser-Compton Gamma-ray Systems

As illustrated in Figure 6, existing laser-Compton-based materials detection studies in the US (LLNL and Duke/HIGS) and Japan (AIST/JAEA) have utilized sources with SSD's in the range of 10 to 100 depending upon gamma-ray energy [7-9]. However, for practical detection and assay, a source SSD in the range of 1,000,000 to 10,000,000 is required. LLNL's approach to meeting these requirements is based upon high-gradient, compact, x-band accelerator technology first pioneered by the

SLAC National Accelerator Laboratory. In order to produce the required beam current (micro-amps), the accelerator must be operated in a macro/micro bunch mode that produces, high-quality, low-emittance (0.1 mm-mrad-scale), electron beams [10] at repetition rates up to ~100 kHz. To test and refine this mode of operation a sub-scale x-band test system has been constructed and is being commissioned (see Figure 7) at LLNL.

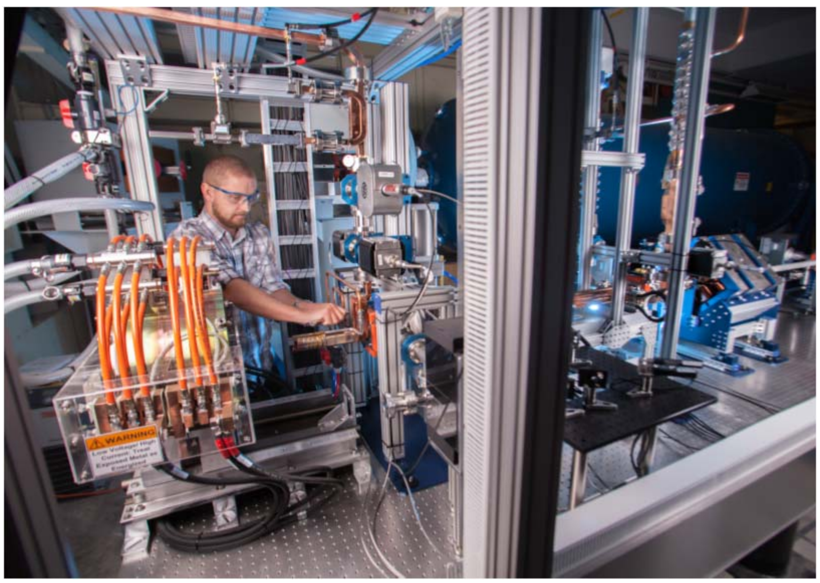

Figure 7. LLNL's compact, x-band accelerator R&D system. Shown are the high brightness photo-gun (below the researcher's hands) and the gun solenoid (left) and x-band accelerator section (right).

The optimized version of this system utilizes a new, mulit-GHz, fiber-laser technology [11] to produce the sub-ps, few micro-Joule UV pulses